Electronic Supplementary information

Ultrafast-laser-treated poly(3,4-ethylenedioxythiophene): poly(styrenesulfonate) electrodes with enhanced conductivity and transparency for semitransparent perovskite solar cells

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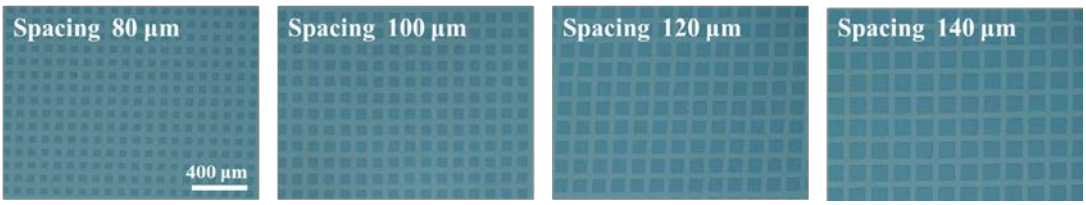


Fig. S1 Optical micrographs of PEDOT:PSS thin films with different laser line-scan spacing at 10 W power.

The trace is the part scanned by the laser. The PEDOT:PSS film processed by the high-power laser is selectively removed, but the surrounding area and the substrate are not destroyed.

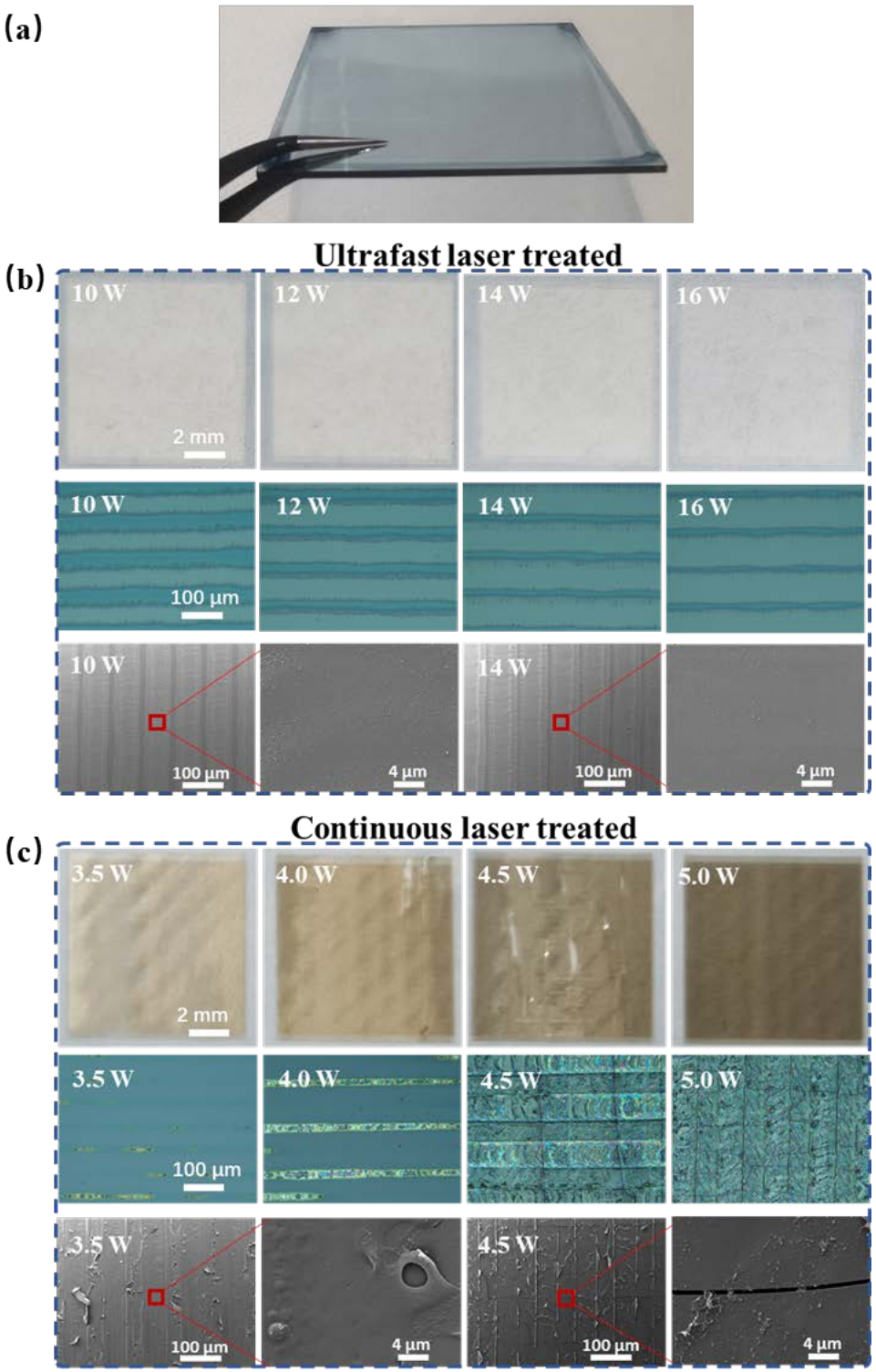


Fig. S2 Selective removal of PEDOT:PSS film on the glass substrate (a) by ultrafast laser (b) and CW laser (c). Camera images, optical microscope images, and SEM images from top to bottom.

The laser line-scan spacing of ultrafast laser processing is 80 μm. The trace is the

laser scanning path. With the increase of laser power, the ablation effect of the laser is more pronounced. Even so, it will not cause damage to the substrate (Fig. S2(b)). With the increase of CW laser power, the film blackens due to intense thermal action (Fig. S2(c)). Cracks appear on the glass substrate due to excessive thermal action.

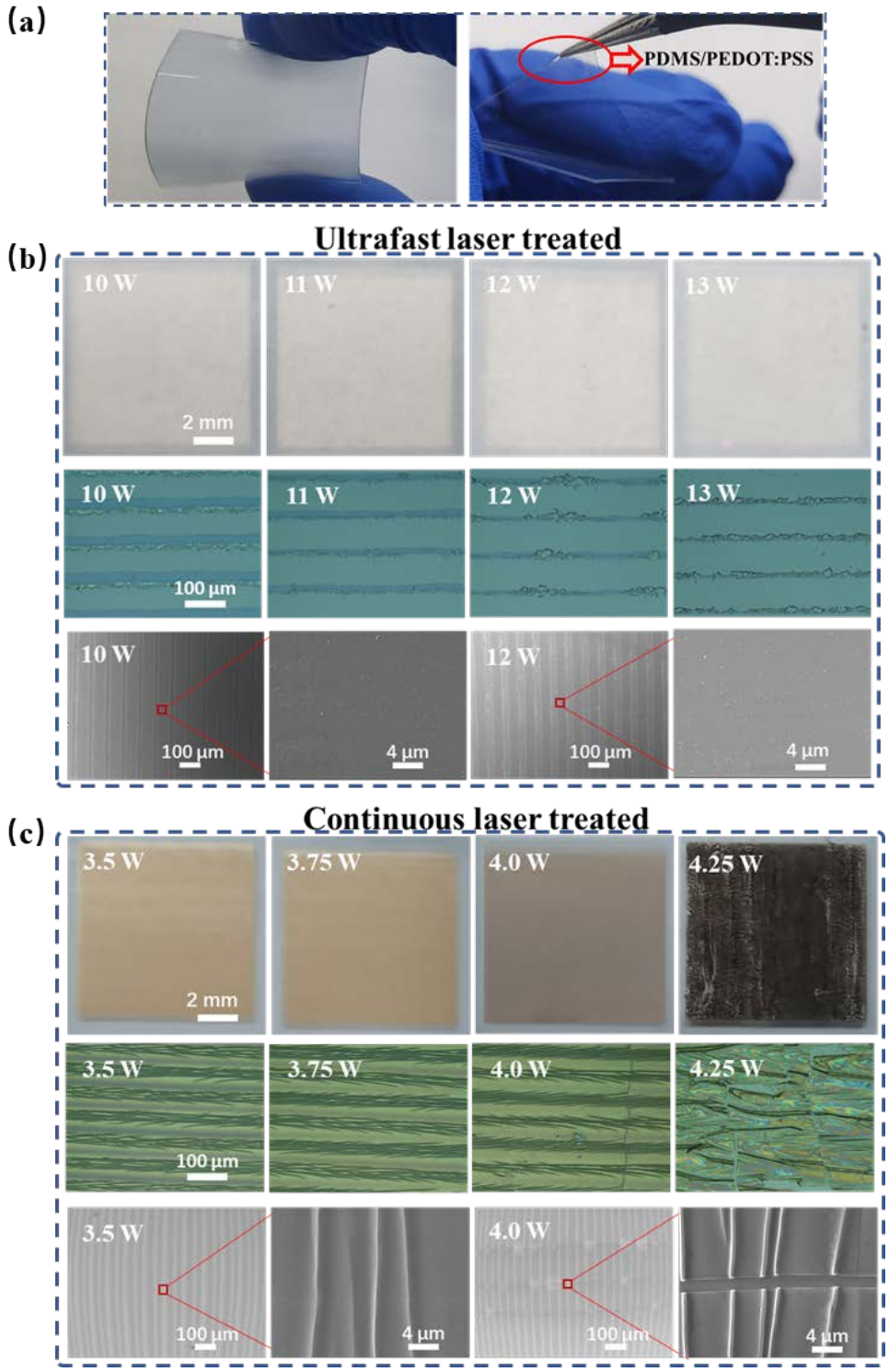


Fig. S3 Selective removal of PEDOT:PSS film on PDMS substrate (a) by ultrafast laser (b) and CW laser (c).

The effect is apparent, consistent with the treatment phenomenon on the glass substrate. It can be seen from Fig. S3(b) that the thermal action of CW laser is intense, which leads to the deformation of the PDMS substrate and the fracture and shedding of PEDOT:PSS.

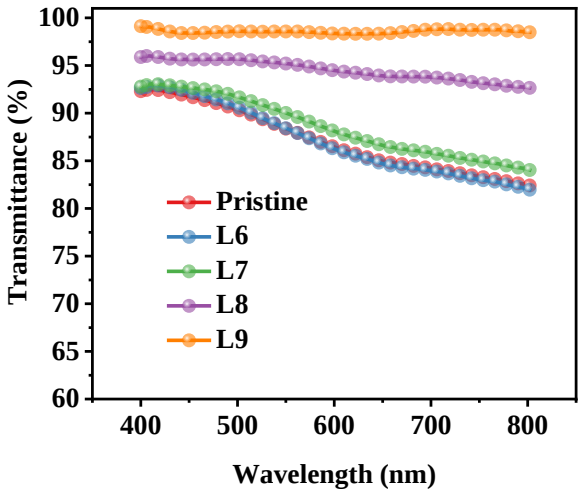


Fig. S4 Transmittance of PEDOT:PSS thin films under different laser power treatments (400–800 nm).

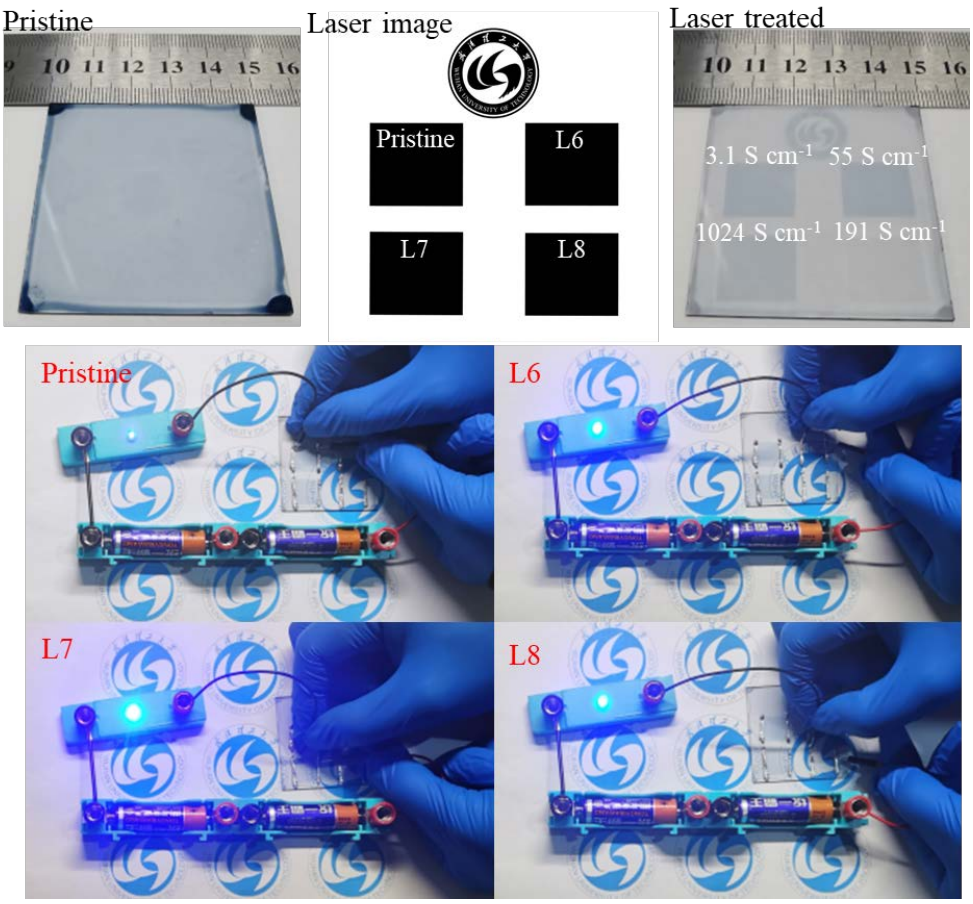


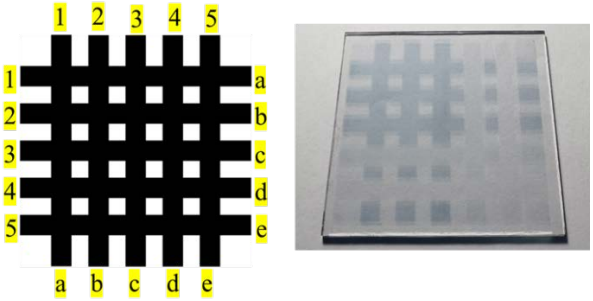
Fig. S5 Led lighting experiment of PEDOT:PSS thin film electrodes treated by different laser. The electrode size is 50 mm × 50 mm.

Line number	A-1	A-2	A-3	A-4	A-5	A-6
Resistance/Ω	145	268	403	547	703	861

Fig. S6 Patterned electrode of 7 W treated thin film.

The electrode pattern shown in the figure uses 7 W power processing. The resistance between the points is shown in the table. Suppose the electrode is connected to the circuit; it can play the role of adjusting the resistance similar to the sliding rheostat.

(a)



(b)

Endpoint	1	2	3	4	5
1	5500	2605	2543	3226	5250
2	4220	891	764	1445	3512
3	4302	837	661	1325	3410
4	4570	1073	885	1544	3635
5	5300	1794	1601	2255	4352

(c)

Line number	1-a	2-b	3-c	4-d	5-e
Power/W	6	6.5	7	7.5	8
R_S**/(Ohm sq^{-1})**	1373	285	85	541	1178

(d)

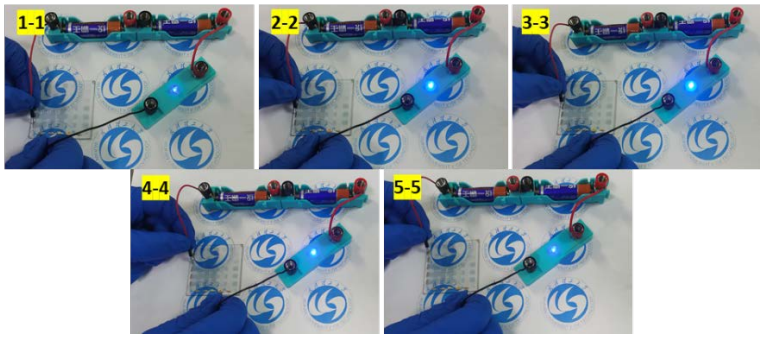


Fig. S7 (a) Patterned electrodes for different laser power treatments. (b) The

resistance between the two endpoints (Ω). (c) The laser processing power and the sheet resistance after the laser processing. (d) Pictures of choosing different endpoints as conductive electrodes to light the LED lamp.

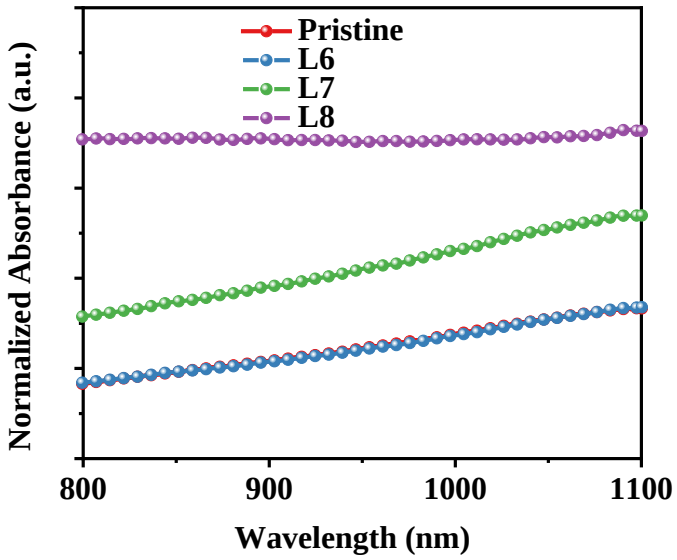


Fig. S8 Absorption of PEDOT:PSS thin films under different power treatments when the wavelength is 800–1100 nm.

The significant increase in the absorption spectrum intensity means that more $PEDOT^{+}$ and $PEDOT^{2+}$ are produced in the film. They indicate that laser treatment improves the oxidation level of the film. This is similar to the PEDOT:PSS film treated by oxidizing acids such as $HClO_4$, HNO_3, H_2SO_4 so on [1, 2]. The oxidation level of the thin film is almost unchanged at the 6 W power treatment. The effect is not apparent when the power is low, consistent with Raman characterization results.

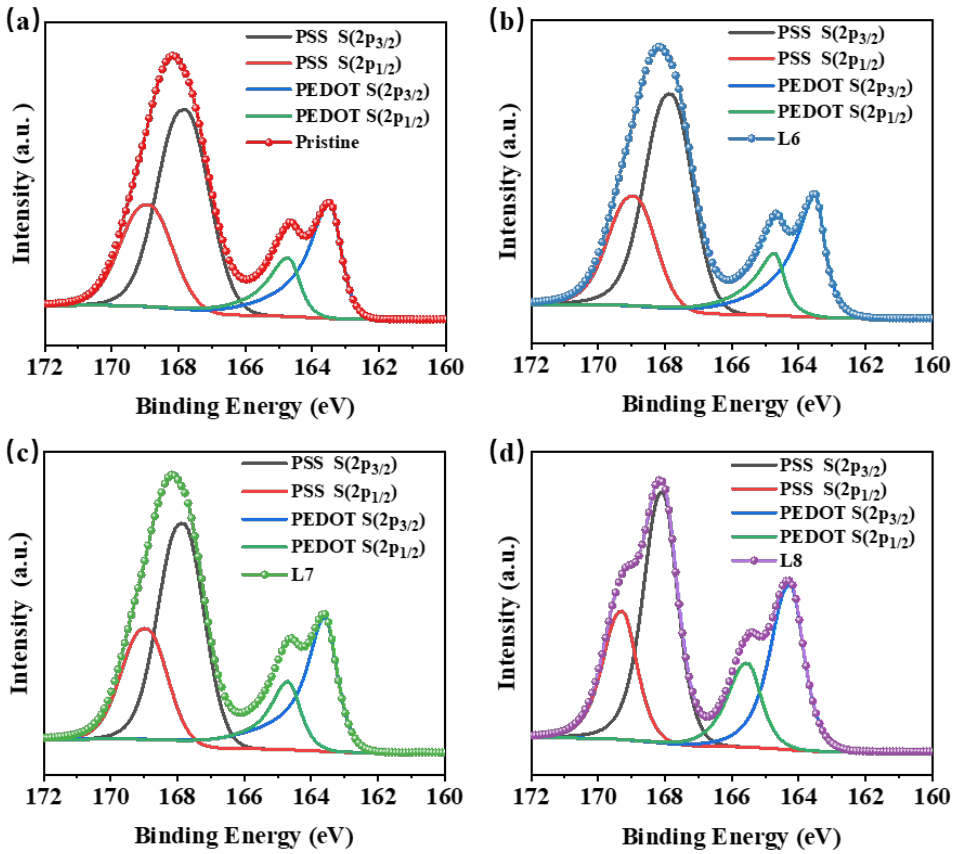


Fig. S9 Deconvolution peak spectrum of XPS of thin films treated with different laser power. (a) Pristine thin films. (b) 6 W treated thin films. (c) 7 W treated thin films. (d) 8 W treated thin films.

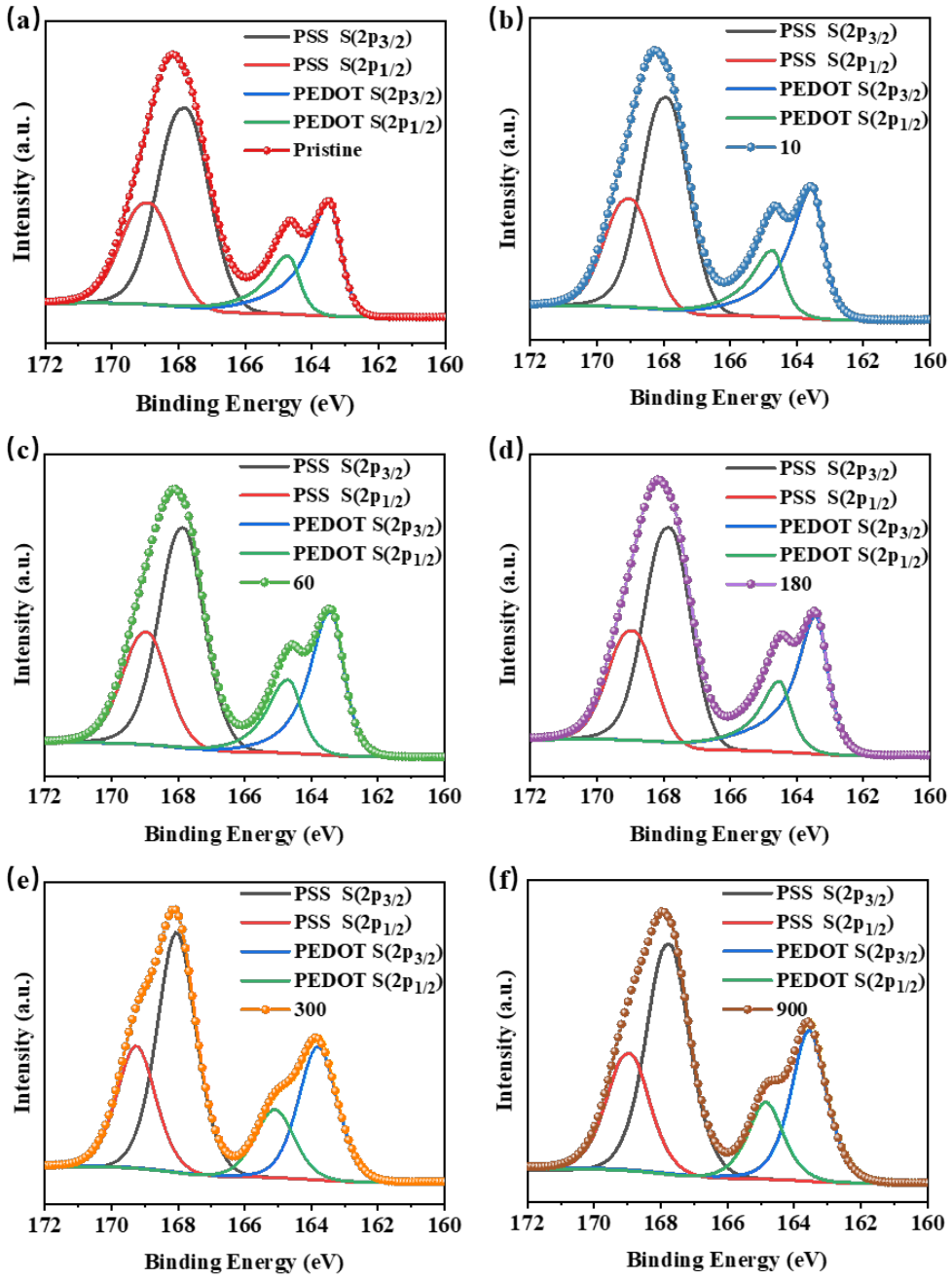


Fig. S10 XPS deconvolution peak spectra of original thin films with different etching time (s). (a) 0 s. (b) 10 s. (c) 60 s. (d) 180 s. (e) 300 s. (f) 900 s.

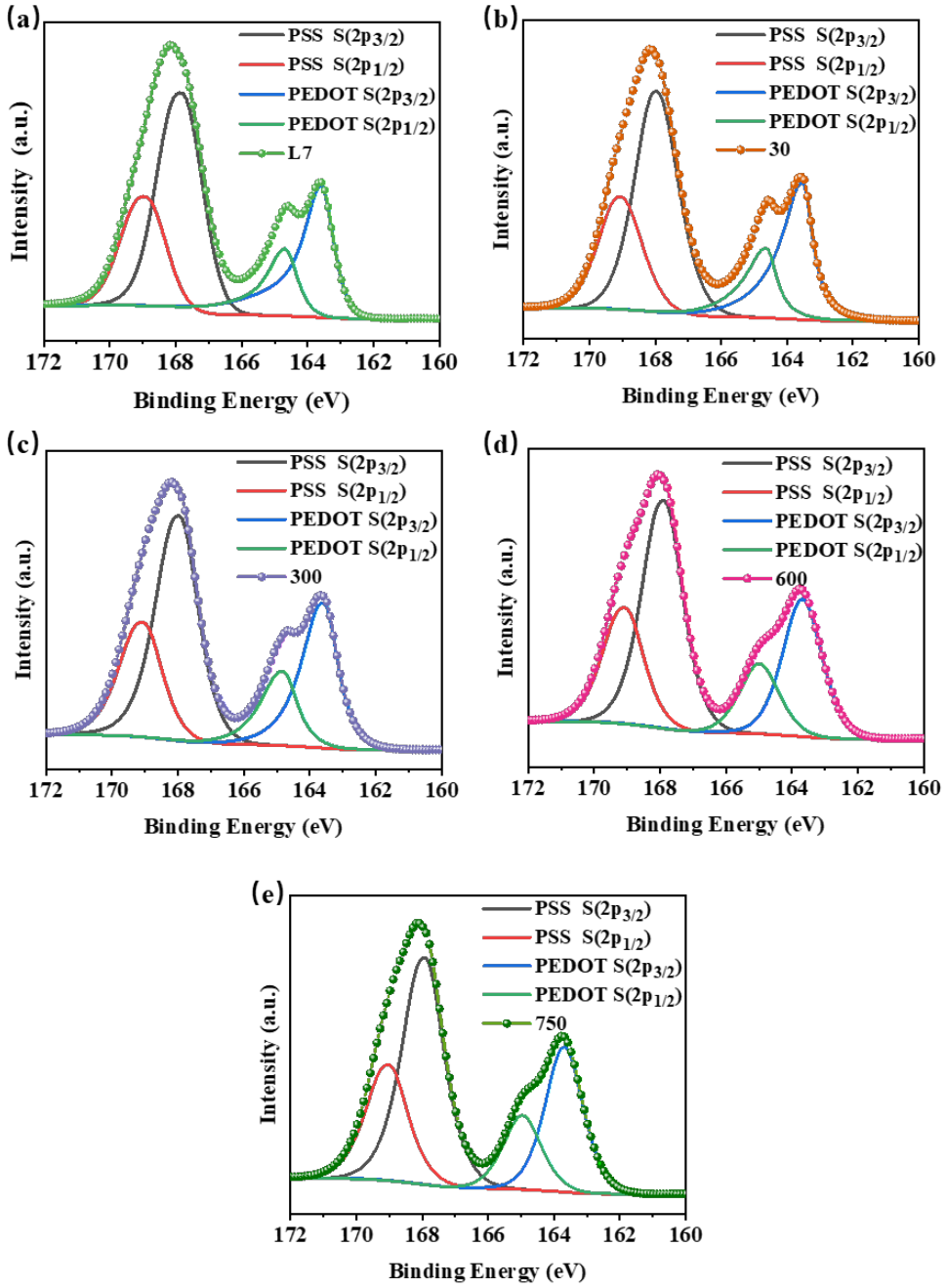


Fig. S11 XPS deconvolution peak spectra of laser-treated thin films with different etching time (s). (a) 0 s. (b) 30 s. (c) 300 s. (d) 600 s. (e) 750 s.

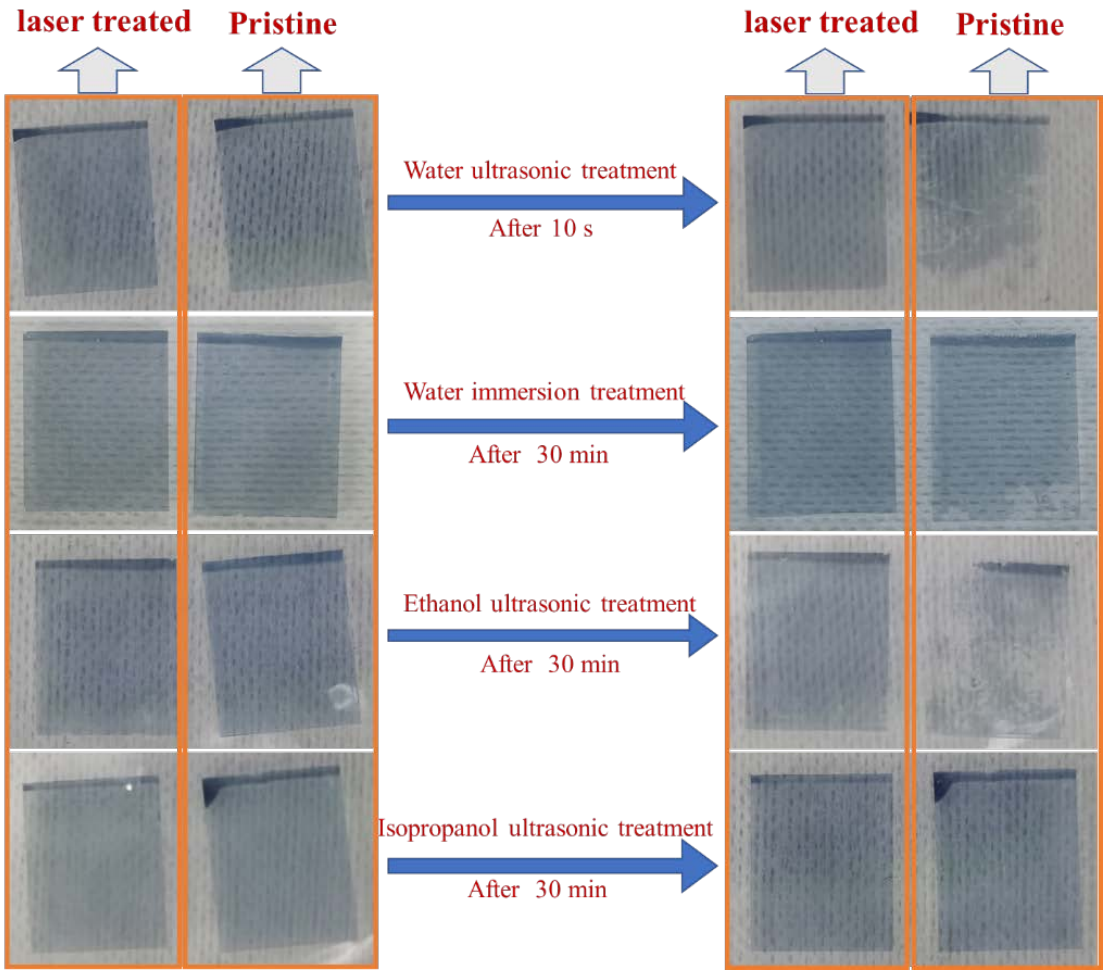
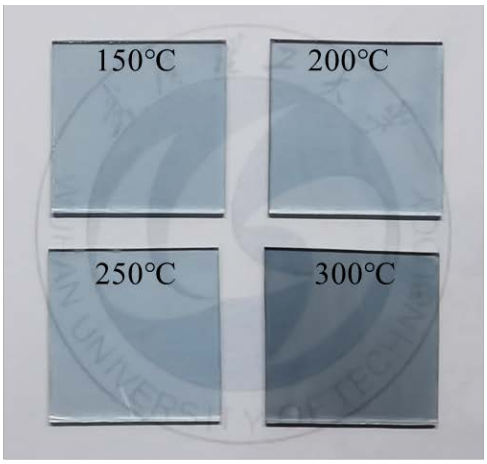


Fig. S12 Stability experiment of laser-treated thin film and original thin film.

Laser treatment will increase the stability of the film in water, ethanol, and isopropanol. After treatment, as shown in the figure, the original film is easier to decompose. The stability of the thin film treated by laser is improved.



Temperature/(℃)	Sheet resistance/(Ohm sq^{-1})
150	20145
200	16124
250	14877
300	--

Fig. S13 Conductivity changes of PEDOT:PSS films under different temperature treatments.

The treatment of PEDOT:PSS films in this figure were as follows: the films prepared by spin coating were heat treated at 150 °C for 10 min, immediately followed by 150 °C, 200 °C, 250 °C, or 300 °C for 10 min. The sheet resistance of the film will decrease a little as the temperature rises. However, the lift of this conductivity is limited.

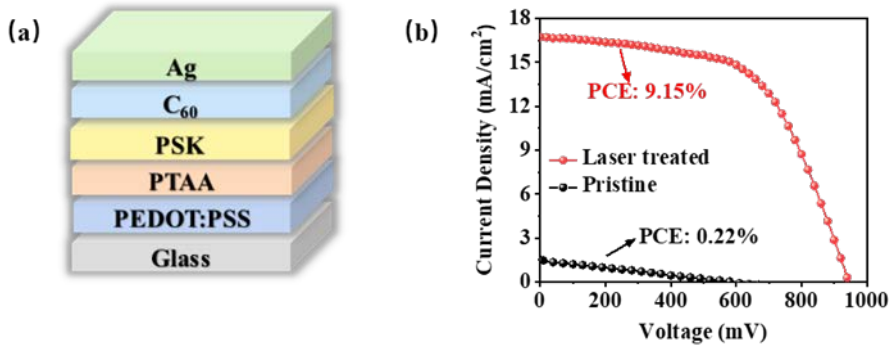


Fig. S14 (a) Structure diagram and (b) current density–voltage curves with the PEDOT:PSS films as the bottom electrodes of perovskite solar cells.

Fig. S14 shows the structure diagram and current density–voltage curves of the perovskite solar cells with the PEDOT:PSS films as the bottom electrodes. The device's champion PCE with laser-processed PEDOT:PSS as the bottom electrode is 9.15%. The V_{OC} is 0.944 V, the J_{SC} density is 16.75 mA cm^{-2}, and the *FF* is 0.578. The PCE with pristine PEDOT:PSS only is 0.22% (Fig. S14(b)), the V_{OC} is 0.59 V, the J_{SC} is 1.50 mA cm^{-2}, and the *FF* is 0.25.

Preparation of perovskite solar cells with PEDOT:PSS as the bottom electrode: Pristine PEDOT:PSS electrodes were prepared by spin coating on clean sodium calcium glass. The prepared pristine PEDOT:PSS film was subjected to 7 W power laser treatment for further use. Poly[bis (4-phenyl)(2,4,6-trimethylphenyl)amine] was prepared by spin coating with the setting of an acceleration rate of 4000 rpm/s at a rate of 4000 rpm for 40 s. It was heat-treated at 100 °C for 10 min after the spin coating was completed. The perovskite component is $Cs_{0.05}(FA_{0.85}MA_{0.15})_{0.95}Pb(I_{0.85}Br_{0.15})_3$. The film was prepared by the spin coating method. The spin coating parameters were set as an acceleration of 1000 rpm/s, a speed of 6000 rpm, and a spin coating time of 30 s, followed by heat

treatment at 120 °C for 50 min. The electron transport material was C_{60}, prepared by vacuum electroless plating, followed by vacuum electroless plating of a 3 nm thick Bathocuproine film. Finally, a 100 nm Ag film was evaporated to complete the preparation of the whole device. Related medicinal products were all purchased from Xi'an Polymer Light Technology Corp. The typical current-density−voltage curves are measured under standard AM 1.5G irradiation at 100 mW cm^{-2}. The size of the mask we used to measure device efficiency is 4 mm × 4 mm, so the effective area of our device is also 4 mm × 4 mm.

Table S1. Report of elemental quantification of XPS when treated with different laser powers.

Peak	Atomic/% -Pristine	Atomic/% -L6	Atomic/% -L7	Atomic/% -L8
C1s	71.3	71.11	69.21	67.69
O1s	21.67	21.76	22.56	23.59
S2p	7.03	7.13	8.23	8.72

Supplementary references

1. Fan Z, Li P, Du D, Ouyang J. Significantly Enhanced Thermoelectric Properties of PEDOT:PSS Films through Sequential Post-Treatments with Common Acids and Bases. Advanced Energy Materials, 2017, 7(8): 1602116
2. Zhang L, Yang K, Chen R, Zhou Y L, Chen S S, Zheng Y J, Li M, Xu C H,

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